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**UTILITY
PATENT APPLICATION
TRANSMITTAL**

(Only for new nonprovisional applications under 37 CFR 1.63(b))

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APPLICATION ELEMENTS

See MPEP chapter 600 concerning utility patent application contents.

ADDRESS TO: Assistant Commissioner for Patents
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1. ☒ Fee Transmittal Form
(Submit an original, and a duplicate for fee processing)
2. ☒ Specification [Total Pages -17]
(preferred arrangement set forth below)
- Descriptive title of the Invention
 - Cross References to Related Applications
 - Statement Regarding Fed sponsored R & D
 - Reference to Microfiche Appendix
 - Background of the Invention
 - Brief Summary of the Invention
 - Brief Description of the Drawings (if filed)
 - Detailed Description
 - Claim(s)
 - Abstract of the Disclosure
3. ☒ Drawing(s) (35 USC 113) [Total sheets -4]
4. ☒ Oath or Declaration [Total Pages - 3]
- a.1. ☒ Newly executed (original or copy)
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- b. ☐ Copy from a prior application (37 CFR 1.63(d))
(for continuation/divisional with Box 17 completed)
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- i. ☐ **DELETION OF INVENTOR(S)**
Signed statement attached deleting inventor(s)
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1.63(d)(2) and 1.33(b).
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The entire disclosure of the prior application, from which
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6. Microfiche Computer Program (Appendix)
7. ☐ Nucleotide and/or Amino Acid Sequence Submission
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- a. ☐ Computer Readable Copy
- b. ☐ Paper Copy (identical to computer copy)
- c. ☐ Statement verifying identity of above copies

ACCOMPANYING APPLICATION PARTS

8. ☒ Assignment Papers (cover sheet & document(s))
9. ☐ 37 CFR 3.73(b) Statement ☐ Power of Attorney
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10. ☐ English Translation Document (if applicable)
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PROCESS FOR INHIBITING A POLYMERIZATION IN A VACUUM SECTION OF AN EASILY POLYMERIZABLE COMPOUND PURIFICATION SYSTEM

BACKGROUND OF THE INVENTION

1. Field of the Invention

The present invention relates to a process for inhibiting a polymerization in a vacuum system, which is used for purifying an easily-polymerization compound such as (meth)acrylic acid and (meth)acrylate under a reduced pressure. The invention also relates to the easily polymerizable compound purification system.

2. Description of the Related Art

In distillation operations of the easily polymerizable compound such as (meth)acrylic acid and (meth)acrylate, polymerization of the easily polymerizable compound must be avoided during distillation. To this end, the pressure inside a distillation column is reduced and the inside temperature is decreased as much as possible, and a polymerization inhibitor such as hydroquinone and phenothiazine, or a gas containing molecular oxygen is used for inhibiting a polymerization.

FIG. 1 is a schematic illustration of a conventional process for the purification of the easily polymerizable compound. This process includes a purifying section which

comprises a distillation column 1 and a condenser 2 and a vacuum section which comprises a steam ejector E1, E2 and E3 for reducing a pressure of a purifying section, a barometric condenser 3, surface condensers 4 and 5 and a ejector seal tank 6.

An easily polymerizable compound is usually distilled in the distillation column 1 under a reduced pressure. The pressure inside the distillation column 1 is controlled by the closing and opening of an air regulating valve 8, while the air regulating valve 8 is controlled on the basis of measurements obtained by a pressure gauge 7. Most of the resulting distilled compound is condensed through a condenser 2. The exhaust gas from the condenser 2 flows into a vacuum section by the aid of a steam ejector E1 which is reducing a pressure of a purifying section. The gas usually contains the easily polymerizable compound which is not condensed through the condenser 2 and other noncondensing gases. The gas and steam through the steam ejector E1 are condensed in a barometric condenser 3 by contacting the gas with a cooling liquid directly. The resulting condensate is withdrawn into a ejector seal tank 6 and a remained gas is sucked into the second steam ejector E2. The gas and steam from the second steam ejector E2 are condensed through a surface condenser 4 and the resulting condensate is withdrawn into the ejector seal tank 6 and a remained gas is sucked into the third steam

ejector E3. In the case of a surface condenser, the gas is condensed without direct contact with a cooling water. The gas from the third ejector E3 is condensed through a surface condenser 5 attached at the downstream side of the third ejector E3. Generally, both the resulting condensate and a remained gas are liquid-sealed by the ejector seal tank 6. In some processes, the gas is not liquid-sealed at a final stage.

FIG. 2 is an illustration of another conventional process for the purification of the easily polymerizable compound. This process includes a purifying section which comprises a distillation column 11 and a condenser 12 and a vacuum section comprising a nash pump 13 as a vacuuming device, a pump 15 and a tank 14 and a cooler 17. The easily polymerizable compound is distilled in a distillation column 11 and the resulting distilled compound is condensed through a condenser 12. The exhaust gas from the condenser 12 flows into a vacuum section by the aid of the nash pump 13 reducing a pressure of a purifying section. The gas usually contains the easily polymerizable compound which is not condensed through the condenser 12 and a noncondensing gas. The gas is usually sealed by the liquid which is circulated from a tank 14 by the nash pump 13 and the pump 15.

According to the conventional process, the gas exhausted from a condenser connected to a distillation column contains an easily polymerizable compound such as (meth)acrylic acid

and (meth)acrylate. The easily polymerizable compound is liable to polymerize, and the resulting polymer clogs a condenser, a liquid ejector, or a nash pump, and the vacuum section is frequently forced to stop.

SUMMARY OF THE INVENTION

An object of the invention is to provide a process which is capable of inhibiting the polymerization in a vacuum section of an easily polymerizable compound purification system.

Another object of the invention is to provide an system which is free from the problems residing in the prior art.

According to an aspect of the invention, a process for inhibiting polymerization in a vacuum section of an easily polymerizable compound purification system comprises the step of

permitting a gas containing an easily polymerizable compound to flow into a gas and liquid contact chamber from a purifying section, the gas and liquid contact chamber being supplied with a liquid containing a polymerization inhibitor.

According to another aspect of the invention, a system for purifying an easily polymerizable compound comprises;

a column for purifying an easily polymerizable compound;

a vacuum portion for vacuuming a gas containing the easily polymerizable compound from the column; and

a liquid supply equipment for supplying a liquid containing a polymerization inhibitor to the vacuum portion to thereby come into contact with the gas.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a schematic illustration of a conventional system for purification of an easily polymerizable compound;

FIG. 2 is a schematic illustration of another conventional system for purification of an easily polymerizable compound;

FIG. 3 is a schematic illustration of a system for purification of an easily polymerizable compound according to an embodiment of the invention; and

FIG. 4 is a schematic illustration of another system for purification of an easily polymerizable compound according to another embodiment of the invention.

DESCRIPTION OF THE PREFERRED EMBODIMENT

The subject-matter of the present invention is that the polymerization in a vacuum section of an easily polymerizable compound purification system can be inhibited by contacting a liquid containing a polymerization inhibitor with a gas containing an easily polymerizable compound in a gas and liquid contact chamber. The liquid is contacted with the compound directly before the compound begins to polymerize in

the vacuum section. The invention has been accomplished based on these findings.

FIG. 3 is a schematic diagram illustrating the invented process and system for the inhibiting the polymerization in the vacuum section of the easily polymerizable compound purification system.

The system shown in FIG. 3 includes a purifying section which comprises a distillation column 1, a condenser 2, a pressure gauge 7, an air regulating valve 8 and pipes connecting them and a vacuum section which comprises a steam ejector E1, E2 and E3 as a vacuuming device, gas and liquid contact chambers 3, 4 and 5, a cooler 22, a pump 21, an ejector seal tank 6 and pipes connecting them. In the system, the pipes must be connected so that a condensed liquid of the gas containing the easily polymerizable compound does not accumulate. For example, a vacuuming nozzle and a exhausting nozzle of the gas and liquid chamber are preferably set horizontally or downward below horizontal. The vacuum section comprises a vacuum portion and a liquid supply equipment.

In this embodiment, the steam ejector and the gas and liquid contact chamber constitutes the vacuum portion and the pump and the ejector seal tank constitutes the liquid supply equipment.

According to this process of the invention, a

polymerization inhibitor is added to a liquid in the ejector seal tank 6, and the liquid containing a polymerization inhibitor is supplied from the ejector seal tank 6 through a line L1 to the inside of the gas and liquid contact chambers 3 and 4 by the aid of a pump 21 to inhibit the polymerization of the easily polymerizable compound in the vacuum section. Examples of the polymerization inhibitors for the process of the invention are hydroquinone, methoquinone, manganese acetate, phenothiazine, nitrosophenol, cupferron, dibutyl dithio carbamic acid copper salt and N-oxyl compounds.

According to the process of the invention, the easily polymerizable compound is distilled in the distillation column 1 under the reduced pressure. Examples of easily polymerizable compounds are (meth)acrylic acids and (meth)acrylates. Examples of (meth)acrylates are methyl(meth)acrylate, ethyl(meth)acrylate, isopropyl(meth)acrylate, n-propyl(meth)acrylate, isobutyl(meth)acrylate, 2-hydroxyethyl(meth)acrylate, hydroxypropyl(meth)acrylate, dialkylaminoethyl(meth)acrylate.

Most of the distilled compound is condensed through a condenser 2. The exhaust gas from the condenser 2 flows into a vacuum section by the aid of a steam ejector E1 which is reducing a pressure of a purifying section. The gas usually contains the easily polymerizable compound which is not

condensed through the condenser 2 and other noncondensing gases. The gas flows into the gas and liquid contact chamber 3 through the steam ejector E1 and is contacted with the liquid containing a polymerization inhibitor which is supplied from the ejector seal tank 6. The gas and liquid contact chamber may be set between the purifying section and the vacuum section. Preferably the gas and liquid contact chamber is a condenser such as a barometric condenser and a surface condenser in a vacuum section.

When the gas and liquid contact chamber is a barometric condenser, the liquid is supplied to cool the barometric condenser and contacted with the compound directly in the condenser so as to inhibit the polymerization of the compound.

When the gas and liquid contact chamber is a surface condenser, the inside surface of the surface condenser is wetted uniformly with the liquid. The liquid is preferably sprayed onto the overall surface of the surface condenser. Such surface condensers include horizontal condensers and vertical condensers, where condensation is performed inside a tube (inside condensation type) or outside the tube (outside condensation type). From the viewpoint of uniform wetting, a vertical condenser is preferred to a horizontal condenser, and an inside condensation type is preferred to an outside condensation type.

When the two or more gas and liquid contact chambers are

included in the vacuum section, preferably the liquid containing the polymerization inhibitor should be supplied to the first gas and liquid contact chamber or to both of the first and the second gas and liquid chambers, because the first gas and liquid chambers usually contains a gas including large amount of the easily polymerizable compounds. In this case, barometric condensers are preferred to surface condensers for the gas and liquid contact chamber.

The vacuum section can include a liquid ejector and/or a nash pump instead of the steam ejector for reducing a pressure of the purifying section. The Liquid ejectors and nash pumps are applied for reducing a pressure of the purifying section in the range of an absolute pressure from 6.7kPa(50mmHg) to 101Kpa(760mmHg). On the other hand, the steam ejector can be employed for almost overall ranges of the pressure of the purifying section by adjusting several stages of ejectors (usually, one to five ejectors).

FIG. 4 is a schematic illustration of another embodiment of the invented process for the distillation of the easily polymerizable compound using a nash pump. This process includes a purifying section which comprises a distillation column 11, a condenser 12, a pressure gauge 16, and a vacuum section which comprises a nash pump 13, a pump 15, a tank 14 and a cooler 17. The vacuum section comprises a vacuum portion and a liquid supply equipment. In this embodiment,

the nash pump 13 constitutes the vacuum portion and the pump 15, the tank 14 and the cooler 17 constitutes the liquid supply equipment. The vacuum portion can employ a combination of the nash pumps, the steam ejectors and the liquid ejectors.

According to this process of the invention, a polymerization inhibitor is added to a liquid in the tank 14, and the liquid is circulated by the nash pump 13 and the pump 15 for reducing a pressure of the purifying section. The gas containing the easily polymerizable compound such as (meth)acrylic acid and (meth)acrylate is sucked into the nash pump 13 and sealed by the liquid circulated from the tank 14 by the aid of the pump 15 so as to inhibit the polymerization of the compound.

When a portion of the liquid is exhausted as a waste gas by the flow rate of the gas, an additional liquid containing the polymerization inhibitor is supplied to compensate for the amount of the waste liquid. Preferably the additional liquid is supplied to avoid increasing the concentration of the compound in the liquid. The waste liquid containing the compound is preferably recycled for recovering the compound and inhibiting a polymerization of a purification section such as an absorbing column, a distillation column and a stripping column.

The invention as thus configured provides a process for inhibiting the polymerization in a vacuum section for

purifying an easily polymerizable compound such as (meth)acrylic acid and (meth)acrylates and a system for purifying an easily polymerizable compound.

The present invention will now be illustrated in more detail with reference to a comparative example and an inventive example below, which are not intended to limit the scope of the invention.

EXAMPLE

A purifying system shown in FIG. 3 includes a purifying section which comprises a distillation column 1, a condenser 2, a pressure gauge 7, an air regulating valve 8 and a vacuum section which comprises a steam ejector E1, E2 and E3 as a vacuuming device, a barometric condenser 3, surface condensers 4 and 5 as a gas and liquid contact chamber, a cooler 22 and a pump 21 and an ejector seal tank 6. Using this system, a liquid containing acrylic acid was put into the distillation column 1, and distillation was continuously performed by operating the system at an overhead absolute pressure of 3.3kPa(25 mm Hg), and distilling off acrylic acid as an overhead product by supplying a liquid containing 100 ppm of hydroquinone as a polymerization inhibitor to the barometric condenser 3 and the surface condenser 4 shown in FIG. 3. As a result, the operation could be continued for one month without a problem. The system was stopped to be inspected, and small amounts of the polymer were observed to attach to

the first barometric condenser and the second surface condenser.

COMPARATIVE EXAMPLE

A purifying system shown in FIG.1 includes a purifying section which comprises a distillation column 1 and a condenser 2 and a vacuum section which comprises a steam ejector E1, E2 and E3 for reducing a pressure of a purifying section, a barometric condenser 3, surface condensers 4 and 5 and a ejector seal tank 6. Using this system, a liquid containing acrylic acid was put into the distillation column 1, and distillation was continuously performed by operating the system at an overhead absolute pressure of 3.3kPa(25 mm Hg), and distilling off acrylic acid as an overhead product without using a polymerization inhibitor. Eight days into the operation, the overhead pressure could not be retained at a constant level of 3.3pKa(25 mm Hg), and the pressure started to increase then. The vacuum section was forced to stop because of the polymerization of acrylic acid.

This application is based on patent application No. 11-156656 filed in Japan, the contents of which is hereby incorporated by reference.

Other embodiments and variations will be obvious to those skilled in the art, and this invention is not to be limited to the specific matters stated above.

WHAT IS CLAIMED IS:

1. A process for inhibiting a polymerization in a vacuum section of an easily polymerizable compound purification system, comprising the step of

permitting a gas containing an easily polymerizable compound to flow into a gas and liquid contact chamber from a purifying section, the gas and liquid contact chamber being supplied with a liquid containing a polymerization inhibitor.

2. A process according to claim 1, wherein the vacuum section comprises one or more gas and liquid contact chambers, and the liquid containing the polymerization inhibitor is supplied to the first gas and liquid contact chamber.

3. A process according to claim 1, wherein the vacuum section comprises two or more gas and liquid contact chambers, and the liquid containing the polymerization inhibitor is supplied to the first and the second gas and liquid contact chambers.

4. A process according to claim 1, wherein the gas and liquid contact chamber is a surface condenser, and the inside surface of the condenser is wetted uniformly with the liquid.

5. A process according to claim 1, wherein the gas and liquid chamber is a barometric condenser, and the liquid serves to cool the barometric condenser.

6. A process according to claim 1, wherein the vacuum section comprises a liquid ejector for reducing a pressure of the purifying section.

7. A process according to claim 1, wherein the vacuum section comprises a nash pump for reducing a pressure of the purifying section.

8. A process according to claim 1, wherein the vacuum section comprises a liquid ejector and a nash pump for reducing a pressure of the purifying section.

9. A process according to claim 1, wherein the easily polymerizable compound is (meth)acrylic acid and/or (meth)acrylate.

10. A process according to claim 1, wherein the polymerization inhibitor is at least one selected from the group consisting of hydroquinone, methoquinone, manganese acetate, phenothiazine, nitrosophenol, cupferron, dibutyl dithio carbamic acid copper salt and N-oxyl compounds.

11. A system for purifying an easily polymerizable compound which comprises;

a column for purifying an easily polymerizable compound;

a vacuum portion for vacuuming a gas containing the easily polymerizable compound from the column; and

a liquid supply equipment for supplying a liquid containing a polymerization inhibitor to the vacuum portion to thereby come into contact with the gas.

12. The system according to claim 11, wherein the vacuum portion comprises at least one or more steam ejectors and at least one or more gas and liquid contact chambers.

13. The system according to claim 11, wherein the gas and liquid contact chamber is a barometric condenser, and the liquid serves to cool the barometric condenser.

14. The system according to claim 11, wherein the gas and liquid contact chamber is a surface condenser, and the inside surface of the surface condenser is wetted with the liquid.

15. The system according to claim 11, wherein the vacuum portion comprises a liquid ejector.

16. The system according to claim 11, wherein the vacuum portion comprises a nash pump.

17. The system according to claim 11, wherein the vacuum portion comprises a liquid ejector and a nash pump.

18. The system according to claim 11, wherein the easily polymerizable compound is (meth)acrylic acid and/or (meth)acrylate.

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ABSTRACT OF THE DISCLOSURE

A process for inhibiting a polymerization of an easily polymerizable compound purification system is disclosed.

According to the present invention, the polymerization of the easily polymerizable compound such as (meth)acrylic acid and (meth)acrylate flowed into a vacuum section can be inhibited by contacting a liquid containing a polymerization inhibitor with the compound directly in the vacuum section.

When the vacuum section includes a gas and liquid contact chamber (usually a condenser), the liquid containing a polymerization inhibitor may be supplied to the chamber. When the vacuum section includes a liquid ejector and/or a nash pump as a vacuuming device, the liquid containing a polymerization inhibitor may be circulated by the liquid ejector and/or the nash pump for reducing a pressure of a purifying section. Examples of easily polymerizable compounds are (meth)acrylic acid and (meth)acrylates. The process of the invention is capable of inhibiting the polymerization of the easily polymerizable compound such as (meth)acrylic acid and (meth)acrylate in a vacuum section of a purifying system. The system of the invention is suitable for purifying the easily polymerizable compound.

FIG.1

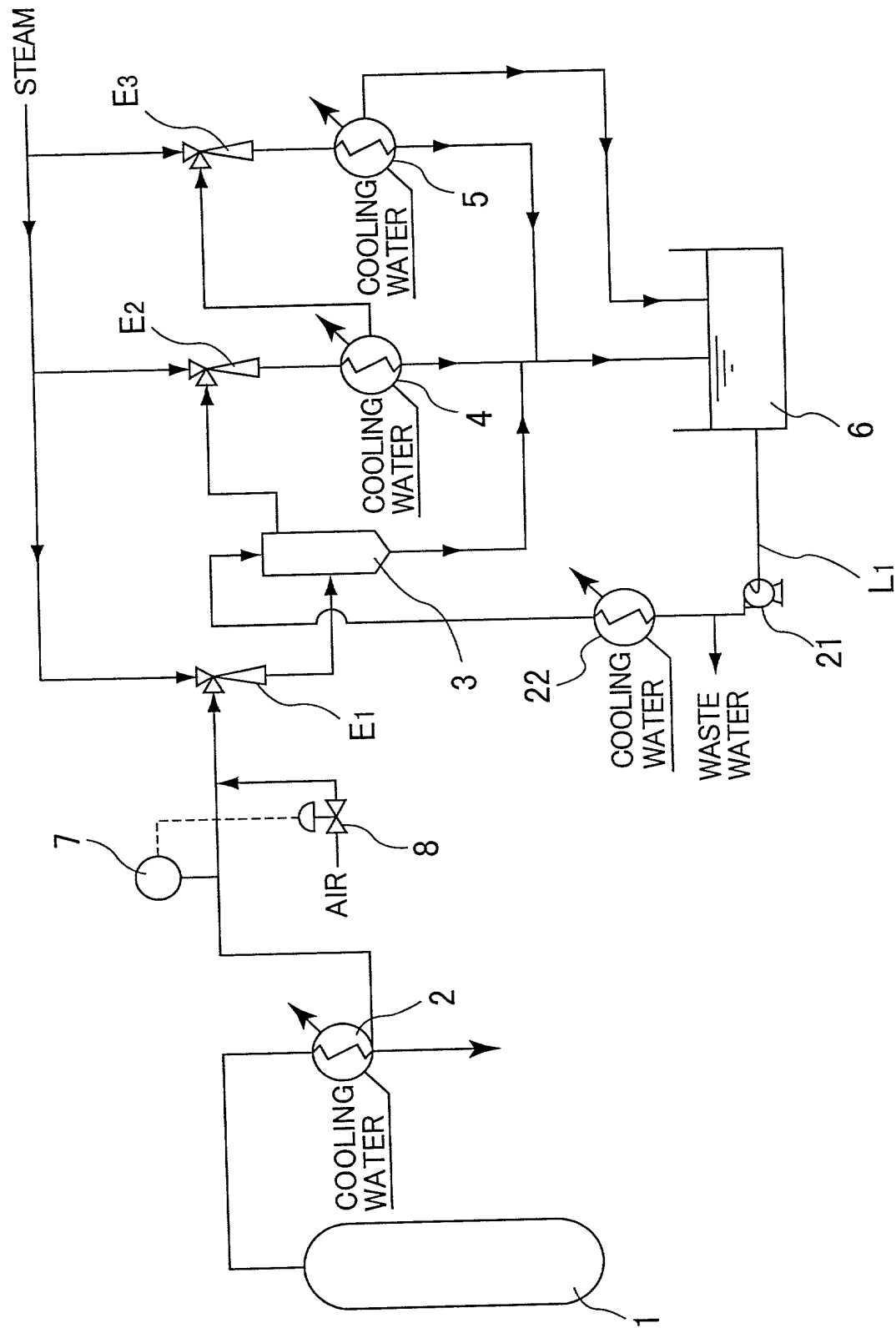


FIG.3

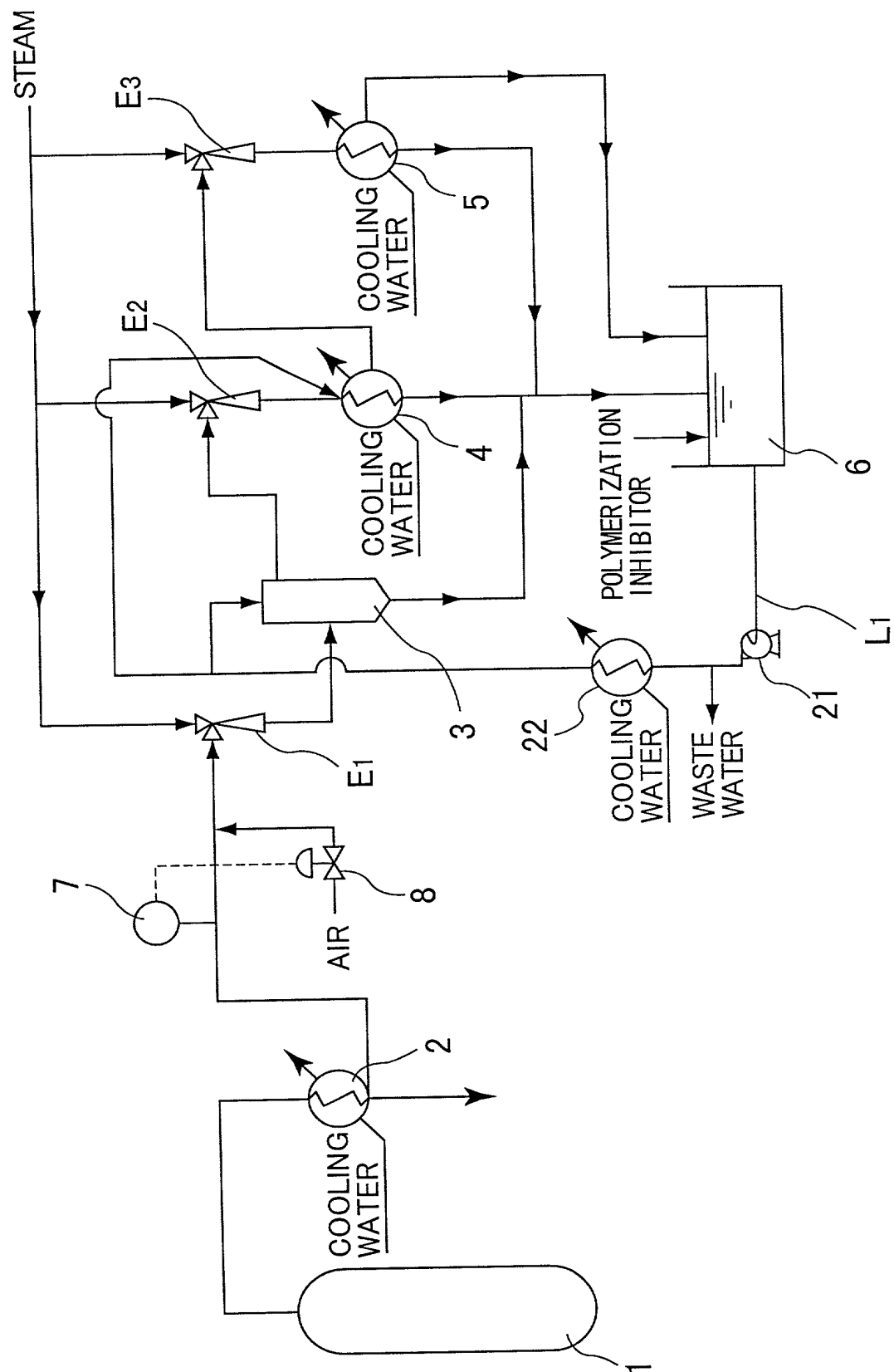
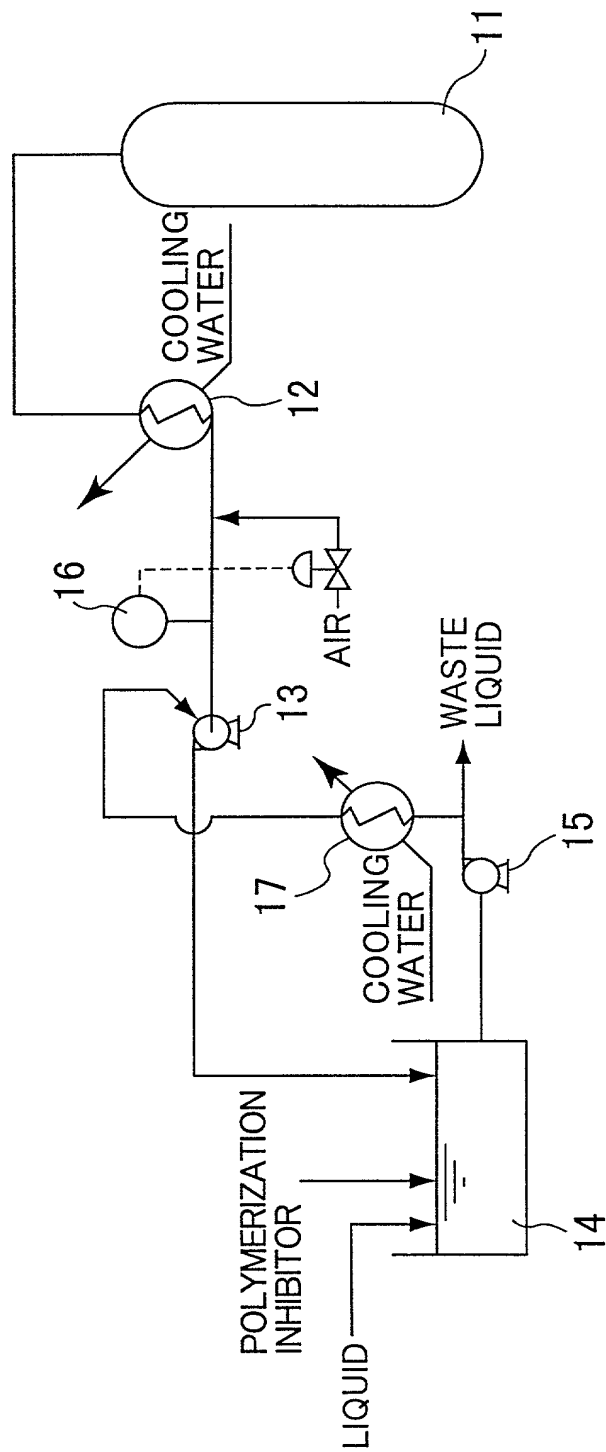


FIG.4



COMBINED DECLARATION AND POWER OF ATTORNEY
FOR PATENT APPLICATION

As a below-named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

PROCESS FOR INHIBITING A POLYMERIZATION IN A VACUUM
SECTION OF AN EASILY POLYMERIZABLE COMPOUND
PURIFICATION SYSTEM

the specification of which

(check one) X is attached hereto.

_____ was filed on _____
as Application Serial No. _____
and was amended on _____
(if applicable)

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, § 1.56.

I hereby claim foreign priority benefits under 35 U.S.C. 119(a)-(d) or 365(b) of any foreign application(s) for patent or inventor's certificate, or 365(a) of any PCT international application which designated at least one country other than the United States of America, listed below and have also identified below, by checking below, any foreign application for patent or inventor's certificate, or of any PCT international application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application(s):

			Priority Claimed	
			<u>X</u>	
(Number)	(Country)	(Day/Month/Year Filed)	Yes	No
<u>11-156656(PAT.)</u>	<u>Japan</u>	<u>03/June/1999</u>		

I hereby claim the benefit under 35 U.S.C. 119(e) of any United States provisional application(s) listed below.

(Application Number)

(Filing Date)

I hereby claim the benefit under 35 U.S.C. 120 of any United States application(s), or 365(c) of any PCT international application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR 1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

(Appl. Serial No.)

(Filing Date)

(Status) (patented, pending, abandoned)

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith. (list name and registration number)

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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